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**(54) Fungicidal compositions**

Fungizide Zusammensetzungen

Compositions fongicides

(84) Designated Contracting States:  
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(56) References cited:  
**EP-A- 0 050 738**                      **EP-A- 0 148 526**  
**EP-A- 0 287 346**                      **EP-A- 0 458 060**  
**EP-A- 0 458 061**                      **GB-A- 2 136 423**  
**US-A- 4 542 146**

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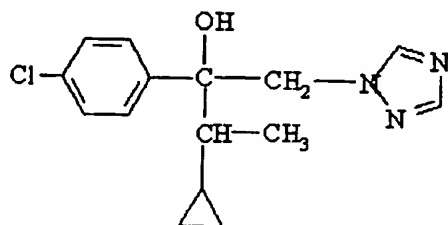
**EP 0 555 186 B1**

## Description

This invention relates to a wood preservative composition and, more specifically, to the use of a wood preservative composition containing Cyproconazole as active ingredient for protecting wood.

Wood is an important resource material in the construction and industries. Wood can, however be susceptible to mold, decay and discoloring due to fungal attack. Various compositions are known for combatting such fungal attacks, including certain triazole compounds such as those disclosed in EP-A-0 050 738, EP-A-0 131 684, EP-A-148 526, EP-A-0 458 060, US-PS 4,548,146. Fungicidal compositions for agricultural use comprising Cyproconazole are known from EP-A-0 287 346 and GB-A-2 136 423. Further, the earlier European Patent Applications EP-A-0 531 837 and EP-A-554 833 are directed to wood preservatives comprising Cyproconazole as active ingredient.

It has now been found that Cyroconazole of the formula (I)



(I)

is particularly effective at combatting various fungi which are known to cause mold, decay and discoloration of wood.

Wood, as used herein, refers to any type of wood material or wood product such as plywood, pressed wood, particle-board, wood chip, pulp or intermediates obtained in papermaking.

The Cyproconazole is commercially available.

The compound of formula (I) for use as wood preservatives is conveniently formulated into compositions comprising a wood preserving or fungicidally effective amount of the compound of formula (I) and an environmentally acceptable carrier for such usage.

The term carrier as used herein means any environmentally acceptable liquid or solid material which may be added to the active constituent to bring it in an easier or improved applicable form, respectively to a usable or desirable strength of activity. It can for example be calcium, magnesium carbonate, xylene or water.

The compositions may also be in the form of dispersible powders or granules and will conveniently comprise a surfactant, e.g. a wetting or dispersing agent to facilitate dispersion in liquids of the powder or granules which may contain also fillers and suspending agents.

The aqueous dispersions or emulsions may be prepared by dissolving the active ingredient in an organic solvent optionally containing wetting, dispersing or emulsifying agents and then adding the mixture to water which may also contain one or more surfactants, such as wetting, dispersing or emulsifying agents. Suitable organic solvents are ethylene dichloride, isopropyl alcohol, propylene glycol, diacetone alcohol, toluene, kerosene, methylnaphthalene, polyethyleneglycol, N-methyl-2-pyrrolidone, mixtures of C9 to C11 fatty alcohols, the xylenes, trichloroethylene, furfuryl alcohol, tetrahydrofurfuryl alcohol and glycol ethers.

Typically, the compositions will be in the form of liquid preparations for use as dips or sprays which are generally aqueous dispersions or emulsions containing the active ingredient in the presence of one or more surfactants e.g. wetting agents, dispersing agents or emulsifying agents. The surfactants may be cationic, anionic or non-anionic, all of which are known in the art.

Suitable anionic agents are soaps, salts of aliphatic monoesters of sulphuric acid and salts of sulphonated aromatic compounds.

Suitable non-ionic agents are the condensation products of ethylene oxide with fatty alcohols or with alkyl phenols. Other non-ionic agents are the partial esters derived from long chain fatty acids and hexitol anhydrides, the condensation products of partial esters with ethylene oxide and the lecithins.

The compositions of the invention may contain further adjuvants including thickening agents, antifoam agents, anti-freeze agents and suspending agents.

Suitable suspending agents are hydrophilic colloids and vegetable gums.

The compositions for use as aqueous dispersions or emulsions are generally supplied in the form of a concentrate containing a high proportion of the active ingredient, the concentrate to be diluted with water before use. The concentrates may conveniently contain up to 95%, suitably 10-85%, for example 25-60% by weight of the active ingredient. After dilution to form aqueous preparations, such preparations may contain varying amounts of the active ingredient

depending upon the type of wood to be treated and the type of fungus, but typically the aqueous preparation will contain from 0.0001% to 10% by weight active ingredient, more typically from 0.001% to 1%.

Methods of applying the compounds to the wood to be treated, such as spraying, dipping, by paint brush, etc., are known to those skilled in the art. Application can be repeated, as necessary.

- 5 The formulations listed below are representative of suitable formulations for use in the invention, and are admixed and agitated in accordance with conventional methods to obtain a wood preservative composition.

#### Formulation 1

- 10 400 g/l cyproconazole  
 55 g/l nonionic polymeric emulsifier blend (e.g. polyalkylene glycol ether/polyoxyethylene alkylaryl ether blend)  
 66 g/l antifreeze (e.g. 1,2 propanediol)  
 3 g/l thickening agent (e.g. xanthane gum)  
 1 g/l bactericide  
 15 4 g/l antifoam agent (e.g. silicon)  
 balance water

#### Formulation 2

- 20 100 g/l cyproconazole  
 57 g/l emulsifier (e.g. a nonylphenolethoxyphosphate)  
 96 g/l solvent (e.g. N-methyl-2-pyrrolidone)  
 balance solvent (e.g. polyethyleneglycol)

#### 25 Formulation 3 (emulsifiable concentrate)

- 100 g/l cyproconazole  
 74 g/l emulsifier (e.g. nonylphenyl-hydroxypoly(oxyethylene)phosphate)  
 92 g/l emulsifier (e.g. alkyl hydroxypoly(oxyethylene)phosphate)  
 30 46 g/l solvent (e.g. hexanol)  
 101 g/l solvent (e.g. N-methyl-2-pyrrolidone)  
 balance solvent (e.g. mixture of C9 to C11 fatty alcohols)

#### Formulation 4 (wetable granule)

- 35 10 % cyproconazole  
 15 % dispersing agent (e.g. sodium lignin sulfonate)  
 75 % carrier (e.g. calcium magnesium carbonate)

#### 40 Test of activity against wood destroying fungi in vitro

- Suspensions containing a test compound of formula I are incorporated into potato dextrose agar (PDA) to produce a series of five concentrations containing 100 ppm, 10 ppm, 1 ppm, 0.1 ppm, 0.01 ppm resp. of active ingredients. The thus obtained agar test compositions are poured into 9-cm petri dishes. After solidification of the medium, each dish is  
 45 inoculated with a mycelial disc (5 mm diameter) taken from the periphery of actively growing colonies on PDA (three replicate dishes per isolate per concentration). After incubation (24°C in darkness, 5-14 days depending on the growth rate of the fungi), colony radii are measured. Percentage growth inhibition is calculated on the basis of treated control plates. The EC90 (effective concentration causing 90 % growth inhibition) is determined on the basis of dose-response curves.

- 50 The compounds of formula (I) are effective in combatting various type of fungi including the following fungi and the symptoms to which they lead.

Fungus class	Species	Sympton
ascomycetes	Sydowia polyspora	dieback/pine
	ceratocysti fagacearum	wilt/oak
	ceratocysti pilifera	blue stain
	Cephaloascus fragrans	mold
	Physalospora rhodina	discoloration
basidiomycetes	Coriolus versicolor	decay
	Poria placenta	decay
	Lentinus lepideus	decay
	Trametes versicolor	decay
	Serpula lacrymans	mold
deuteromycetes	Coniophora putanea	decay
	Gloeophyllum trabeum	decay
	Aspergillus niger	discoloration
	Phialophora fastigiata	discoloration
	Alternaria alternata	discoloration
	Rhinocladiella atrovirens	discoloration
	Gliocladium roseum	mold
	Aureobasidium pullulans	discoloration
	Trichoderma viride	decacy
	Sphaeropsis sapinea	dieback/conifers
	Pencillium expansum	mold

#### Fungicidal activity

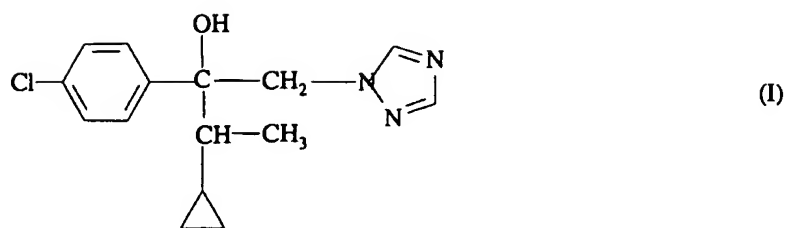
The compound cyproconazole when tested against a variety of fungal diseases demonstrates particularly good activity against basidiomycetes including the fungi *Coriolus versicolor*, *Poria placenta*, *Serpula lacrymans*, *Coniophora puteana*, *Gloeophyllum trabeum*, *Lentinus lepideus* and *Trametes versicolor*.

Cyproconazole is particularly effective against *Poria placenta*, *Lentinus lepideus* and *Trametes versicolor*.

#### Claims

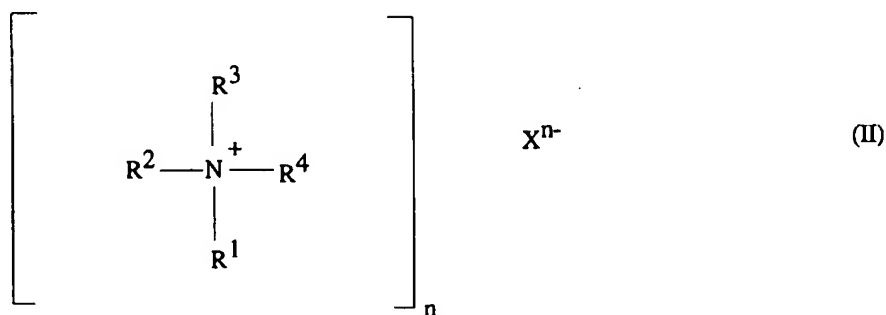
Claims for the following Contracting States : AT, BE, DK, ES, GR, NL, SE

1. Use of a composition comprising a wood preserving effective amount of cyproconazole of the formule I



15 and an environmentally acceptable carrier as wood preserving agent, with the proviso that the composition does not contain

a) an ammonium salt of the formula II



wherein R<sup>1</sup>-R<sup>4</sup> are the same or different organic substituents having up to 18 carbon atoms which are bonded through a C-N- bond, or wherein three of the radicals R<sup>1</sup>-R<sup>4</sup> together with the N-atom form a heteroaromatic system, X<sup>n-</sup> is the anion of an inorganic or organic acid, and n is 1, 2 or 3,

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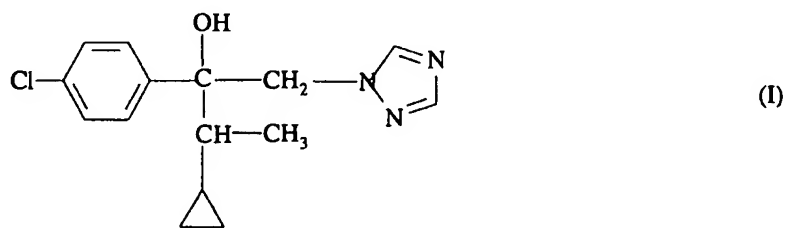
b) methyl methoxyimino- $\alpha$ -(o-tolyloxy)-o-tolylacetate.

2. Use of a composition according to claim 1, wherein the composition comprises additionally a surfactant.

40 3. The use of a composition according to claim 1 for the protection of wood against mold, decay and discoloring due to fungal attack.

#### Claims for the following Contracting States : IE, LU

45 1. Use of a composition comprising a wood preserving effective amount of cyproconazole of the formula I

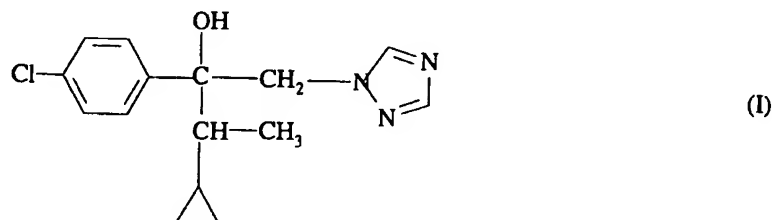


and an environmentally acceptable carrier, as wood preservative agent.

2. Use of a composition according to claim 1, wherein the composition comprises additionally a surfactant.
3. A method for preserving wood against mold decay and discoloring due to fungal attack, comprising applying to the surface of said wood a wood preserving effective amount of cyproconazole of formula I according to claim 1.
4. The use of a composition according to claim 1 for the protection of wood against mold, decay and discoloring due to fungal attack.

**Claims for the following Contracting State : PT**

1. Use of a composition comprising a wood preserving effective amount of cyproconazole of the formula I



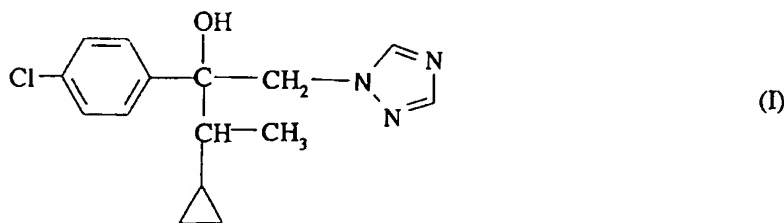
and an environmentally acceptable carrier as wood preserving agent, with the proviso, that the composition does not contain methyl methoxyimino- $\alpha$ -(o-tolyloxy)-o-tolylacetate.

2. Use of a composition according to claim 1, wherein the composition comprises additionally a surfactant.
3. A method for preserving wood against mold decay and discoloring due to fungal attack, comprising applying to the surface of said wood a wood preserving effective amount of cyproconazole of formula I according to claim 1.
4. The use of a composition according to claim 1 for the protection of wood against mold, decay and discoloring due to fungal attack.

**Patentansprüche**

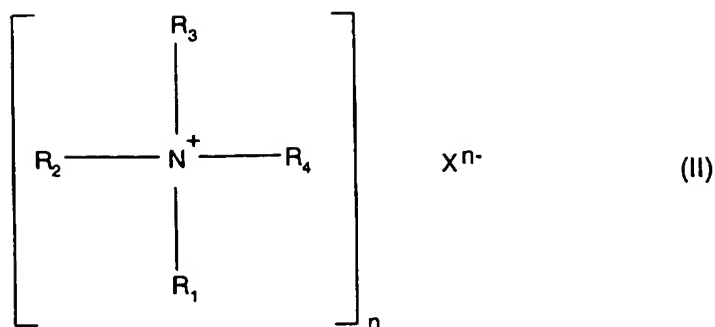
**Patentansprüche für folgende Vertragsstaaten : AT, BE, DK, ES, GR, NL, SE**

1. Verwendung einer Zusammensetzung enthaltend eine holzkonservierend wirksame Menge von Cyproconazole der Formel I



und einen für die Umwelt annehmbaren Träger als Holzschutzmittel, mit der Massgabe, dass die Zusammensetzung nicht enthält

- a) ein Ammoniumsalz der Formel II



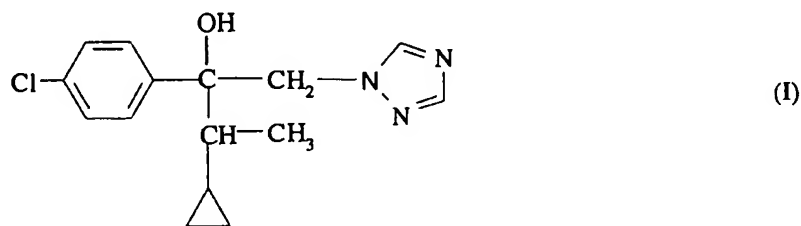
worin  $R_1$  bis  $R_4$  gleiche oder verschiedene organische Substituenten mit bis zu 18 Kohlenstoffatomen sind, welche durch eine C-N-Bindung gebunden sind, oder worin 3 der Radikale  $R_1$  bis  $R_4$  zusammen mit dem N-Atom ein heteroaromatisches System bilden,  $X^{n-}$  ein Anion einer anorganischen oder organischen Säure ist und  $n$  1, 2 oder 3 ist,

b) Methyl-methoximino- $\alpha$ -(*o*-tolylloxy)-*o*-tolylacetat.

2. Verwendung einer Zusammensetzung gemäss Anspruch 1 worin die Zusammensetzung zusätzlich ein oberflächenaktives Mittel enthält.
3. Ein Verfahren zum Schützen von Holz gegen Schimmel, Fäulnis und Verfärbung durch Pilzbefall, umfassend die Anwendung einer holzkonservierend wirksamen Menge von Cyproconazole der Formel I gemäss Anspruch auf die Oberfläche des besagten Holzes.

#### Patentansprüche für folgende Vertragsstaaten : IE, LU

1. Verwendung einer Zusammensetzung enthaltend eine holzkonservierend wirksame Menge von Cyproconazole der Formel I

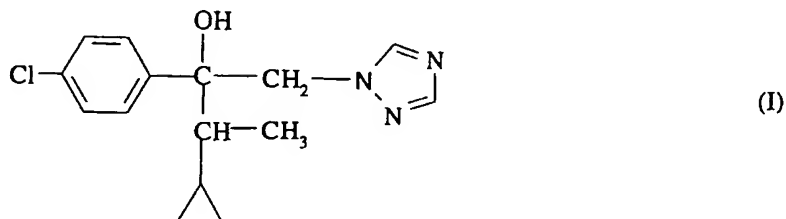


und einen für die Umwelt annehmbaren Trägerstoff, als Holzkonservierungsmittel.

2. Verwendung einer Zusammensetzung gemäss Anspruch 1, worin die Zusammensetzung zusätzlich ein oberflächenaktives Mittel enthält.
3. Ein Verfahren zum Schützen von Holz gegen Schimmel, Fäulnis und Verfärbung durch Pilzbefall, umfassend die Anwendung einer holzkonservierend wirksamen Menge von Cyproconazole der Formel I gemäss Anspruch 1 auf die Oberfläche des besagten Holzes.
4. Verwendung einer Zusammensetzung gemäss Anspruch 1 zum Schutz von Holz gegen Schimmel, Fäulnis und Verfärbung durch Pilzbefall.

Patentansprüche für folgende Vertragsstaat : PT

1. Verwendung einer Zusammensetzung enthaltend eine holzkonservierend wirksame Menge von Cyproconazole der Formel I



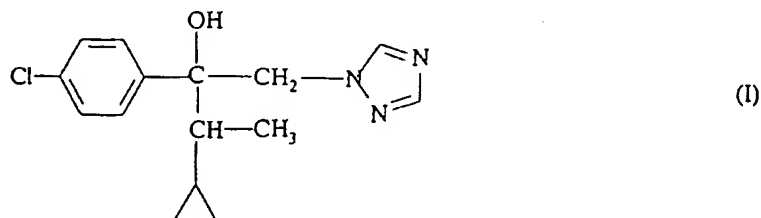
und einen für die Umwelt annehmbaren Träger als Holzkonservierungsmittel, mit der Massgabe, dass die Zusammensetzung nicht Methyl-methoximino- $\alpha$ -(o-tolyloxy)-o-tolylacetat enthält.

2. Verwendung einer Zusammensetzung gemäss Anspruch 1, worin die Zusammensetzung zusätzlich ein oberflächenaktives Mittel enthält.
3. Ein Verfahren zum Schützen von Holz gegen Schimmel, Fäulnis und Verfärbung durch Pilzbefall, umfassend die Anwendung einer holzkonservierend wirksamen Menge von Cyproconazole der Formel I gemäss Anspruch 1 auf die Oberfläche des besagten Holzes.
4. Verwendung einer Zusammensetzung gemäss Anspruch 1 zum Schutz von Holz gegen Schimmel, Fäulnis und Verfärbung durch Pilzbefall.

Revendications

Revendications pour les Etats contractants suivants : AT, BE, DK, ES, GR, NL, SE

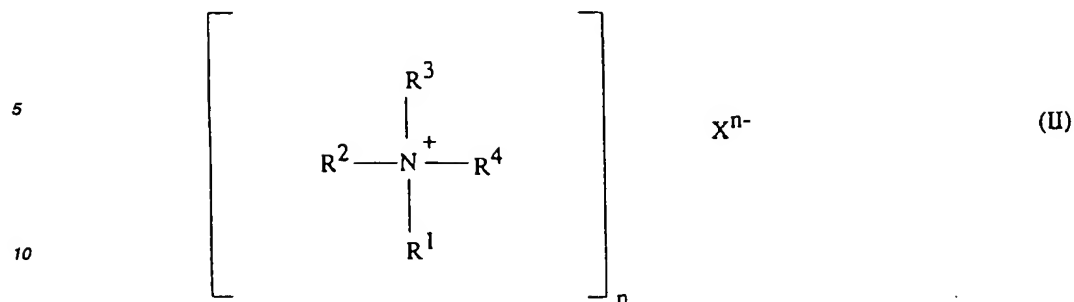
1. Utilisation d'une composition comprenant une quantité efficace pour protéger le bois de cyproconazole de formule I



et un support acceptable pour l'environnement comme agent de protection du bois, avec la condition que la composition ne contienne pas

- a) de sel d'ammonium de formule II





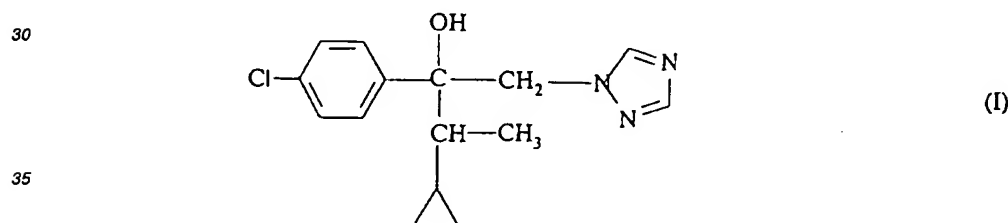
15 dans laquelle R<sup>1</sup> à R<sup>4</sup> sont des substituants organiques identiques ou différents contenant jusqu'à 18 atomes de carbone qui sont liés par une liaison C-N, ou dans laquelle 3 des restes R<sup>1</sup> à R<sup>4</sup> forment ensemble, avec l'atome d'azote, un système hétéroaromatique, X<sup>n-</sup> signifie l'anion d'un acide minéral ou organique et n signifie 1, 2 ou 3,

b) de méthoxyimino- $\alpha$ -(o-tolyloxy)-o-tolylacétate de méthyle.

- 20 2. Utilisation d'une composition selon la revendication 1, où la composition comprend également un tensio-actif.
3. Utilisation d'une composition selon la revendication 1 pour la protection du bois contre la moisissure, la décomposition et la décoloration provoquées par une attaque fongique.

25 **Revendications pour les Etats contractants suivants : IE, LU**

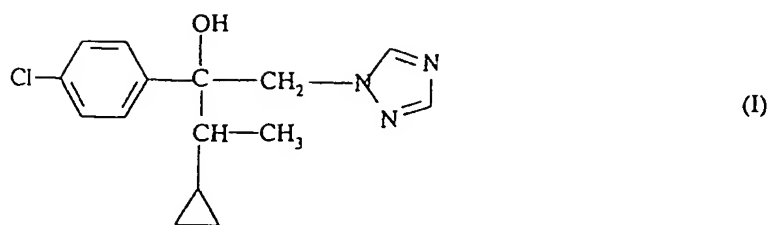
1. Utilisation d'une composition comprenant une quantité efficace pour protéger le bois de cyproconazole de formule I



- 40 et un support acceptable pour l'environnement, comme agent de protection du bois.
2. Utilisation d'une composition selon la revendication 1, où la composition comprend également un tensio-actif.
3. Une méthode de protection du bois contre la moisissure, la décomposition et la décoloration provoquées par une attaque fongique, comprenant l'application à la surface dudit bois d'une quantité efficace pour protéger le bois de cyproconazole de formule I selon la revendication 1.
4. Utilisation d'une composition selon la revendication 1 pour la protection du bois contre la moisissure, la décomposition et la décoloration provoquées par une attaque fongique.

50 **Revendications pour l'Etat contractant suivant : PT**

1. Utilisation d'une composition comprenant une quantité efficace pour protéger le bois de cyproconazole de formule I



et un support acceptable pour l'environnement comme agent de protection du bois, avec la condition que la composition ne contienne pas de méthoxyimino- $\alpha$ -(o-tolyloxy)-o-tolylacétate de méthyle.

- 15
2. Utilisation d'une composition selon la revendication 1, où la composition comprend également un tensio-actif.
  3. Une méthode de protection du bois contre la moisissure, la décomposition et la décoloration provoquées par une attaque fongique, comprenant l'application à la surface dudit bois d'une quantité efficace pour protéger le bois de
  - 20 cyproconazole de formule I selon la revendication 1.
  4. Utilisation d'une composition selon la revendication 1 pour la protection du bois contre la moisissure, la décomposition et la décoloration provoquées par une attaque fongique.
- 25